

# Dynamic steady-state of periodically-driven quantum systems

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Using the density matrix formalism, we prove an existence theorem of the periodic steady-state for an arbitrary periodically-driven system. This state has the same period as the modulated external influence, and it is realized as an asymptotic solution ( $t \rightarrow +\infty$ ) due to relaxation processes. The presented derivation simultaneously contains a simple computational algorithm non-using both Floquet and Fourier theories, i.e. our method automatically guarantees a full account of all frequency components. The description is accompanied by the examples demonstrating a simplicity and high efficiency of our method. In particular, for three-level  $\Lambda$ -system we calculate the lineshape and field-induced shift of the dark resonance formed by the field with periodically modulated phase. For two-level atom we obtain the analytical expressions for signal of the direct frequency comb spectroscopy with rectangular light pulses. In this case it was shown the radical dependence of the spectroscopy lineshape on pulse area. Moreover, the existence of quasi-forbidden spectroscopic zones, in which the Ramsey fringes are significantly reduced, is found. The obtained results have a wide area of applications in the laser physics and spectroscopy, and they can stimulate the search of new excitation schemes for atomic clock. Also our results can be useful for many-body physics.

PACS numbers: 42.62.Fi, 42.62.Eh, 45.30.+s, 03.65.-w, 05.10.-a

For the last several decades a rapid scientific-and-technological progress was substantially connected with an expansion of the lasers and laser technologies at different areas of the science, engineering and industry. In this process the laser physics, nonlinear optics and laser spectroscopy take a special place. Many impressive successes in these directions are due to the theoretical support, motivation and interpretation of experimental researches. In this context a paramount importance has the formulation of mathematical models (equations) and finding of their solutions, which adequately describe the physical picture of investigated problems. As an example, for atomic mediums the density matrix formalism is the most widespread approach describing the atom-field interaction and different relaxation processes (spontaneous, collisional, etc.). Especial significance has so-called steady-state, which arises under the interaction of quantum system with stationary external fields. During long time this state plays a key role for theoretical description of the basic problems in the laser physics and spectroscopy [1–3].

However, in the last years the devices, in which different parameters of electromagnetic fields are periodically modulated, have gotten a great importance. First of all, it is so-called frequency comb-generators using the periodic pulse modulation of the laser field. Such sources of the pulse radiation are actively used now in the modern atomic clocks for frequency measurements [4, 5], and also they have promising perspectives for the direct frequency comb spectroscopy [6–8]. Also it is widely used now the phase (frequency) and/or amplitude pe-

riodic modulation of the laser field to form the reference resonances for small-size atomic clocks and magnetometers [9–14]. Other promising and interesting direction in atomic clocks and magnetometers is connected with periodically modulated polarization of the laser field [15–20]. Of late years the understanding of periodically-driven systems is also one of the most active areas of research in many-body physics (e.g., see [21–25]). In all above examples the *standard* conception of steady-state is inapplicable, generally speaking, due to a regular time variation of external influence.

In this paper, we generalize the steady-state conception for an arbitrary quantum system under any periodic external influence. In this way we prove the following existence theorem: if the coefficients of density matrix dynamic equations have the period  $T$ , then the periodic solution with the same period  $T$  exists always. Due to the relaxation processes this solution is realized as an asymptotics ( $t \rightarrow +\infty$ ) and therefore can be characterized as periodic steady-state. It is found the simple algorithm, which allows us directly to construct this solution independently of initial conditions and without the use of both Floquet and Fourier theories. Our approach considerably simplifies the analysis of the atom-field interaction regardless of periodic modulation character: from smoothly harmonic type to the ultrashort pulses. As a bright example, for two-level atoms we analytically calculate the signal of the direct frequency comb spectroscopy formed by the periodic sequence of rectangular pulses. The obtained results have great perspectives in the laser physics and spectroscopy, and they can stimulate the search of new excitation schemes for atomic clock.

The general mathematical formulation of the problem is following. For an arbitrary quantum system (e.g., atoms) let us consider the density matrix  $\hat{\rho}(t)$  and its

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normalization condition, which in the basis of states  $\{|j\rangle\}$  can be presented as:

$$\hat{\rho}(t) = \sum_{a,b} |a\rangle \rho_{ab}(t) \langle b|; \quad \text{Tr}\{\hat{\rho}(t)\} = \sum_j \rho_{jj}(t) = 1, \quad (1)$$

where  $\rho_{ab}(t)$  are the matrix elements. The dynamic equation for density matrix we reformulate to the vector form:

$$\partial_t \vec{\rho}(t) = \hat{L}(t) \vec{\rho}(t); \quad \text{Tr}\{\hat{\rho}(t)\} = (\vec{n}, \vec{\rho}(t)) = 1, \quad (2)$$

where the linear operator  $\hat{L}(t)$  describes the interaction with external fields and different relaxation processes (spontaneous, collisional, etc.), the column-vector  $\vec{\rho}(t)$  is formed by the matrix elements  $\rho_{ab}(t)$  using some definite rule. As an example, for two-level system with basis states  $|1\rangle$  and  $|2\rangle$  there are four matrix elements  $\rho_{ab}(t)$  ( $a, b=1,2$ ), which can be ordered as following vector:

$$\vec{\rho}(t) = \begin{pmatrix} \rho_{11}(t) \\ \rho_{12}(t) \\ \rho_{21}(t) \\ \rho_{22}(t) \end{pmatrix}. \quad (3)$$

Besides we define the supplementary column-vector  $\vec{n}$  permitting to express the value  $\text{Tr}\{\hat{\rho}(t)\}$  as the dot product in Eq. (2), where we use the standard definition of the dot product for arbitrary complex-valued vectors:  $(\vec{x}, \vec{y}) = \sum_m x_m^* y_m$ . The vector  $\vec{n}$  is formed in the following way: for the positions corresponding to the diagonal elements  $\rho_{jj}(t)$  [as components of the vector  $\vec{\rho}(t)$ ] the vector  $\vec{n}$  has the value 1, and it equals to 0 for all other positions. In particular, for two-level system according to the definition (3) we obtain:

$$\vec{n} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \quad (4)$$

The normalization condition in (2) implies the degeneracy of equations [right-hand members,  $\hat{L}(t)\vec{\rho}(t)$ ] that corresponds to the following expression:

$$(\vec{n}, \hat{L}(t)\vec{\rho}) = 0 \quad (5)$$

for an arbitrary vector argument  $\vec{\rho}$ . Note that the existence of the time-independent vector  $\vec{n}$  is a base point for the next reasonings.

Let us suppose an existence of the time period  $T$  in the operator  $\hat{L}(t)$ :

$$\hat{L}(t+T) = \hat{L}(t). \quad (6)$$

In this case, as it will be shown below, the equation (2) always has a periodic solution with the same period  $T$ :

$$\vec{\rho}(t+T) = \vec{\rho}(t). \quad (7)$$

To begin the prove of this theorem, we assume that at some instant of time  $t_1$  we have an arbitrary vector  $\vec{\rho}(t_1)$ .

Then, in accordance with Eq. (2), for other instant of time  $t_2$  we can write:

$$\vec{\rho}(t_2) = \hat{A}(t_2, t_1) \vec{\rho}(t_1), \quad (8)$$

where the two-time evolution operator  $\hat{A}(t_2, t_1)$  is determined by the matrix  $\hat{L}(t)$ . Note that in the case of periodicity condition (6) we have the following relationship

$$\hat{A}(t_2 + T, t_1 + T) = \hat{A}(t_2, t_1) \quad (9)$$

for arbitrary  $t_1, t_2$ .

However, before the consideration of periodic case, let us prove several general statements. For this purpose, we multiply the Eq. (8) by the vector  $\vec{n}$  as the dot product:

$$(\vec{n}, \vec{\rho}(t_2)) = (\vec{n}, \hat{A}(t_2, t_1) \vec{\rho}(t_1)) = (\hat{A}^\dagger(t_2, t_1) \vec{n}, \vec{\rho}(t_1)), \quad (10)$$

where  $\hat{A}^\dagger(t_2, t_1)$  is the Hermitian conjugate operator to the  $\hat{A}(t_2, t_1)$ . Because of  $(\vec{n}, \vec{\rho}(t_2)) = (\vec{n}, \vec{\rho}(t_1))$ , from Eq. (10) we obtain:

$$(\vec{n}, \vec{\rho}(t_1)) = (\hat{A}^\dagger(t_2, t_1) \vec{n}, \vec{\rho}(t_1)) \quad (11)$$

for arbitrary  $\vec{\rho}(t_1)$ . This formula directly implies

$$\hat{A}^\dagger(t_2, t_1) \vec{n} = \vec{n}, \quad (12)$$

i.e. the operator  $\hat{A}^\dagger(t_2, t_1)$  always has the eigenvector with the real eigenvalue 1. Therefore, the same eigenvalue 1 also exists for the direct operator  $\hat{A}(t_2, t_1)$ , i.e. always there is the eigenvector  $\vec{r}(t_2, t_1)$ , which satisfies the equation:

$$\hat{A}(t_2, t_1) \vec{r}(t_2, t_1) = \vec{r}(t_2, t_1). \quad (13)$$

In contrast to Eq. (12), the eigenvector  $\vec{r}(t_2, t_1)$  depends on  $t_2, t_1$  in the general case.

Let us return to the case of periodicity (6). Consider the  $\vec{\rho}(t)$  at arbitrary instant of time  $t$ . In conformity with Eq. (8), the vector  $\vec{\rho}(t+T)$  is determined as

$$\vec{\rho}(t+T) = \hat{A}(t+T, t) \vec{\rho}(t). \quad (14)$$

Supposing the existence of the periodic solution  $\vec{\rho}(t+T) = \vec{\rho}(t)$ , it follows from Eq. (14) that this solution satisfies the equation:

$$\vec{\rho}(t) = \hat{A}(t+T, t) \vec{\rho}(t); \quad (\vec{n}, \vec{\rho}(t)) = 1, \quad (15)$$

which always has nonzero solution due to the above proven statement of existence of eigenvector with eigenvalue 1 for the operator  $\hat{A}(t_2, t_1)$  for arbitrary  $t_1, t_2$  [see Eq. (13)]. Using the Eqs. (9) and (14),(15), it can be easily shown that  $\vec{\rho}(t+lT) = \vec{\rho}(t)$  ( $l = \pm 1, \pm 2, \dots$ ). Taking into account an arbitrariness of the time  $t$  in the Eqs. (14),(15), we can assert that the existence theorem of the periodic solution (7) is proven.

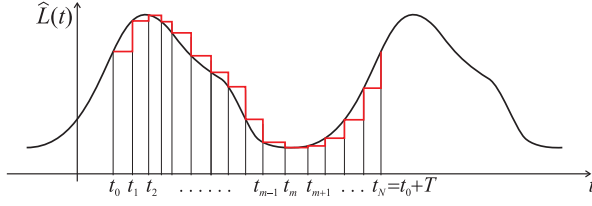


FIG. 1: The partition of the time interval  $[t_0, t_0 + T]$  at  $N$  subintervals and symbolic approximation of the dependence  $\hat{L}(t)$  by the step function (red step line).

Due to the relaxation processes there is an unique (in the majority of cases) periodic solution, which is realized during the time evolution as an asymptotic state ( $t \rightarrow +\infty$ ) independently of initial conditions (similarly to the well-known case of stationary field (18) – see below). The solution (7) can be called as *dynamic* steady-state because of its dependence on  $t$ .

However, taking into account a mathematical generality, one can theoretically assume the possibility of several solutions  $\{\vec{\rho}_1(t), \vec{\rho}_2(t), \dots, \vec{\rho}_Q(t)\}$  for Eq. (15), when the eigenvalue 1 for the matrix  $\hat{A}(t+T, t)$  is degenerated. In this case the general periodic solution has a form of superposition:

$$\vec{\rho}(t) = \sum_{q=1}^Q \alpha_q \vec{\rho}_q(t); \quad (\vec{n}, \vec{\rho}_q(t)) = 1; \quad \sum_{q=1}^Q \alpha_q = 1, \quad (16)$$

where the number set  $\{\alpha_1, \dots, \alpha_Q\}$  for the asymptotic solution ( $t \rightarrow +\infty$ ) will depend on initial conditions (for the dynamical consideration).

Let us show that the found solution (15) is a generalization of well-known steady-state for the constant operator  $\hat{L}$  in Eq. (2):

$$\hat{L}\vec{\rho}_{\text{st-st}} = 0; \quad \partial_t \vec{\rho}_{\text{st-st}} = 0; \quad (\vec{n}, \vec{\rho}_{\text{st-st}}) = 1. \quad (17)$$

It is obviously that the stationary case can be considered as periodic case with an arbitrary value of the period  $T$ . Therefore, according to the above analysis, the state  $\vec{\rho}_{\text{st-st}}$  also must satisfy the Eq. (15) for arbitrary  $t, T$ . Indeed, in the case of  $\hat{L} = \text{const}$  we have (for arbitrary  $t, T$ ):

$$\hat{A}(t+T, t) = e^{\hat{L}T} = 1 + \sum_{k=1}^{+\infty} \frac{1}{k!} T^k \hat{L}^k. \quad (18)$$

Taking into account Eq. (18), we obtain the perfect compliance with Eq. (15):  $\hat{A}(t+T, t)\vec{\rho}_{\text{st-st}} = \vec{\rho}_{\text{st-st}}$ .

Let us compare our results with the general Floquet theory (e.g., see [26]). On this theory the solutions of the differential equation system (2) with periodic coefficients can be presented as following form:

$$\vec{\rho}(t) = \hat{D}(t)e^{\hat{K}t}\vec{a}, \quad (19)$$

where the matrix  $\hat{D}(t) = \hat{D}(t+T)$  is periodic,  $\hat{K}$  is a certain constant matrix connected with initial matrix  $\hat{L}(t)$ ,

$\vec{a}$  is an arbitrary constant vector. To have the periodic solution the matrix  $\hat{K}$  must satisfy certain criteria. From this viewpoint, it is most probably that in the present paper we have proved the existence of the eigenvector  $\vec{a}_0$  with eigenvalue 0 for the matrix  $\hat{K}$ :  $\hat{K}\vec{a}_0 = 0$ . In this case, taking into account  $e^{\hat{K}t}\vec{a}_0 = \vec{a}_0$ , from Eq. (19) we find the following solution:

$$\vec{\rho}(t) = \hat{D}(t)e^{\hat{K}t}\vec{a}_0 = \hat{D}(t)\vec{a}_0, \quad (20)$$

which obviously is periodic [due to the periodicity of  $\hat{D}(t)$ ]. We believe that exactly this scenario is realized for density matrix equation. However, this problem requires an additional mathematical investigation.

Note that some authors supposed (without proof) the existence of the periodic solution (7) for some certain problems. In this case they usually used the Fourier analysis for numerical calculations (e.g., see [10, 27]). However, as far as we know, the general mathematical proof of the existence theorem was not previously presented in scientific literature. At the same time a special interest has the direct and simple method, which allows us to calculate the periodic solution on the base of Eq. (15) without Fourier expansion, i.e. our approach automatically takes account of all frequency components. It can radically simplify the calculations for periodically-driven systems.

Let us describe one of possible numerical algorithms based on Eq. (15). We consider an arbitrary periodic dependence of the operator  $\hat{L}(t)$  (see Fig.1). For instance, under atom-field interaction such a dependence can be produced by the modulation of the field parameters (amplitude, phase, polarization, etc.). The selected time interval  $[t_0, t_0 + T]$  we divide into  $N$  small subintervals, where  $t_N = t_0 + T$ . The character of partition (uniform or nonuniform discrete mesh) and number of subintervals are determined in conformity with studied problem. The dependence  $\hat{L}(t)$  we will approximate by step function (see Fig. 1), where the matrix  $\hat{L}(t)$  has the constant value  $\hat{L}(t_{m-1})$  inside of subinterval  $(t_{m-1}, t_m]$ . In this case the vector  $\vec{\rho}(t_0)$  in initial point  $t_0$  is determined by Eq. (15), where the evolution operator  $\hat{A}(t_0 + T, t_0)$  has the form of an ordered product of the matrix exponents:

$$\hat{A}(t_0 + T, t_0) = \prod_{m=1}^{m=N} e^{(t_m - t_{m-1})\hat{L}(t_{m-1})} = e^{(t_N - t_{N-1})\hat{L}(t_{N-1})} \dots e^{(t_1 - t_0)\hat{L}(t_0)}. \quad (21)$$

The vectors  $\vec{\rho}(t_m)$  in other points of the interval  $[t_0, t_0 + T]$  are determined by the recurrence relation:

$$\vec{\rho}(t_m) = e^{(t_m - t_{m-1})\hat{L}(t_{m-1})}\vec{\rho}(t_{m-1}). \quad (22)$$

Below we give two examples, which demonstrate the simplicity and high efficiency of our method. First example is the so-called dark resonance, which is formed in three-level  $\Lambda$ -system by bichromatic field (see Fig. 2).

This resonance occurs when a difference between the optical frequencies ( $\omega_1 - \omega_2$ ) is varied near the transition frequency between the lower energy levels  $|1\rangle$  and  $|2\rangle$ :  $\omega_1 - \omega_2 \approx \Delta$ . Currently, such resonances at the hyperfine structure of the alkali metal atoms (first of all, for  $^{87}\text{Rb}$  and  $^{133}\text{Cs}$ ) underlie the chip-scale atomic clocks, which have a great practical importance. At the same time the harmonic phase modulation (frequency deviation) of miniature semiconductor lasers (so-called VICSEL) is ordinarily used in practice. In this case the field can be written as

$$E(t) = E_0 e^{-i[\omega t + \varphi(t)]} + c.c.; \quad \varphi(t) = A \sin(\nu t), \quad (23)$$

where  $E_0$  is the field amplitude,  $\omega$  is the central frequency of laser,  $A$  and  $\nu$  are the amplitude and frequency of phase modulation, respectively. It is obvious that this field is polychromatic, in which the frequency difference between the adjacent components equals to  $\nu$ . The phase modulation at full- or half of the hyperfine splitting is usually used:  $\nu \approx \Delta$  or  $\Delta/2$ . However, the dark resonance can be observed in the more general case of  $\nu \approx \Delta/l$  ( $l=1,2,3,\dots$ ).

For theoretical description of this problem the Fourier expansion of the field (23) is traditionally used. In this case, to solve the density matrix equation one can extract two resonant components (e.g., see [10]), which are involved in the absorption and form the dark resonance per se. While the remaining frequency components are taken into account only from the viewpoint of field-induced shifts of the clock transition. It is evident, that such approach is quite cumbersome and incomplete. In our method the Fourier expansion is not used and we directly calculate the time-dependent periodic  $\hat{\rho}(t)$ . In the resonance approximation the equations for the density matrix components  $\rho_{jk}(t)$  have the following form:

$$\begin{aligned} [\partial_t + \Gamma - i\delta_1 - i\varphi'(t)]\rho_{31} &= i\Omega_1(\rho_{11} - \rho_{33}) + i\Omega_2\rho_{21} \\ [\partial_t + \Gamma - i\delta_2 - i\varphi'(t)]\rho_{32} &= i\Omega_2(\rho_{22} - \rho_{33}) + i\Omega_1\rho_{12} \\ [\partial_t + G_0 - i\Delta]\rho_{12} &= i(\Omega_1^*\rho_{32} - \rho_{13}\Omega_2) \\ [\partial_t + G_0]\rho_{11} &= \gamma\rho_{33}/2 + G_0\text{Tr}\{\hat{\rho}\}/2 + i(\Omega_1^*\rho_{31} - \rho_{13}\Omega_1) \\ [\partial_t + G_0]\rho_{22} &= \gamma\rho_{33}/2 + G_0\text{Tr}\{\hat{\rho}\}/2 + i(\Omega_2^*\rho_{32} - \rho_{23}\Omega_2) \\ [\partial_t + G_0 + \gamma]\rho_{33} &= i(\Omega_1\rho_{13} - \rho_{31}\Omega_1^*) + i(\Omega_2\rho_{23} - \rho_{32}\Omega_2^*) \\ \rho_{jk} &= \rho_{kj}^* \quad (j, k = 1, 2, 3); \quad \text{Tr}\{\hat{\rho}\} = \rho_{11} + \rho_{22} + \rho_{33} = 1. \end{aligned} \quad (24)$$

Here  $\delta_{1,2} = (\omega - \omega_{1,2}^{(0)})$  are one-photon detunings,  $\Omega_1 = d_{31}E_0/\hbar$  and  $\Omega_2 = d_{32}E_0/\hbar$  are the Rabi frequencies ( $d_{31}$  and  $d_{32}$  are the reduced matrix elements of the dipole moment for the respective transitions),  $\gamma$  is decay rate of excited level  $|3\rangle$ ,  $\Gamma$  is the total decoherence rate (spontaneous, collision, time-of-flight) of optical transitions  $|1\rangle \rightarrow |3\rangle$  and  $|2\rangle \rightarrow |3\rangle$ ,  $G_0$  is relaxation rate of lower energy levels to the equilibrium isotropic state:  $(|1\rangle\langle 1| + |2\rangle\langle 2|)/2$ . Note, if we delete the derivative by phase modulation  $\varphi'(t)$ , then the equations (24) formally coincide with the description of interaction between monochromatic field (at the frequency  $\omega$ ) and three-level  $\Lambda$ -system.

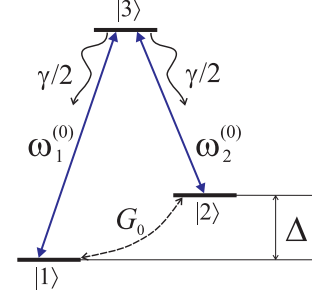


FIG. 2: Three-level system where the dark resonance can be formed.

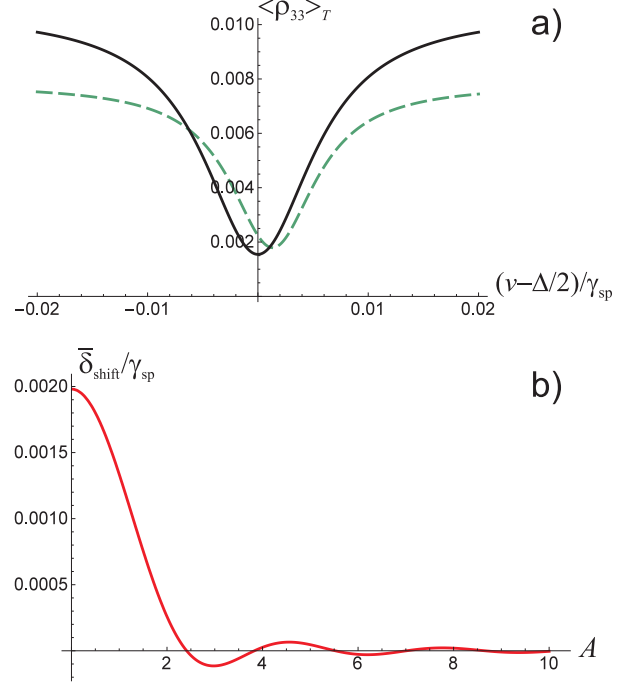


FIG. 3: a) The dark resonance lineshape in the case when  $\nu$  is varied near  $\Delta/2$  for different values of the phase modulation amplitude  $A$ :  $A=1.0$  (green dashed line) and  $A=2.4$  (solid black line); b) The dependence of the dark resonance position (relative to  $\Delta/2$ ) on parameter  $A$ . Calculations are done for  $\Omega_1/\gamma = \Omega_2/\gamma = 1$ ;  $G_0/\gamma = 1.5 \times 10^{-3}$ ;  $\Gamma/\gamma = 50$ ;  $\Delta/\gamma = 1000$ ,  $\delta_2 = -\delta_1 = \Delta/2$ .

In particular, we have calculated by our method the resonance lineshapes as dependencies on the frequency of phase modulation  $\nu \approx \Delta/2$  for different values of amplitude  $A$  (see Fig. 3a). The taken parameters are typical for atomic cells with vapor of  $^{87}\text{Rb}$  in buffer gas. Fig. 3b displays the dependence of field shift of dark resonance  $\bar{\delta}_{\text{shift}}$  (with respect to  $\Delta/2$ ) on modulation amplitude  $A$ . There is the value set of  $A$ , for which the field shift vanishes (see also [28]). In Fig. 4 the similar graphics are shown for the case of phase modulation frequency  $\nu \approx \Delta/4$ . From Fig. 4a it is seen that the amplitude of dark resonance can be comparable with the case of  $\nu \approx \Delta/2$  (compare with Fig. 3a).

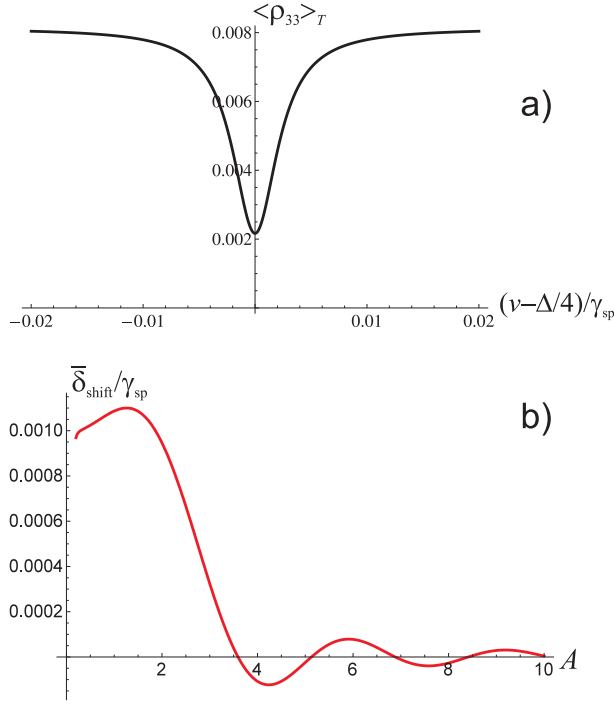


FIG. 4: a) The dark resonance lineshape in the case when  $\nu$  is varied near  $\Delta/4$  for the phase modulation amplitude  $A=3.59$ ; b) The dependance of the dark resonance position (relative to  $\Delta/4$ ) on parameter  $A$ . Calculations are done for  $\Omega_1/\gamma=\Omega_2/\gamma=1$ ;  $G_0/\gamma=1.5 \times 10^{-3}$ ;  $\Gamma/\gamma=50$ ;  $\Delta/\gamma=1000$ ,  $\delta_2=-\delta_1=\Delta/2$ .

To show other examples, let us consider two-level system (with unperturbed frequency  $\omega_0$ ) interacting with the field at the frequency  $\omega$ :  $E(t)=\text{Re}\{\mathcal{E}(t)e^{-i\omega t}\}$ , where the amplitude modulation  $\mathcal{E}(t)$  has the form of rectangular-pulse periodic sequence (see Fig. 5a), which can be considered as limiting case of the Ramsey spectroscopy. In this case the field has an equidistant spectrum  $\omega_m=\omega+m f_r$  ( $m=0,\pm 1,\pm 2,\dots$ ) (so-called frequency comb), where  $f_r=2\pi/T$  is the repetition frequency, and spectrum width (with respect to the central frequency  $\omega$ ) is determined by the pulse duration as  $1/\tau$ . The dynamic equations for density matrix components are given by:

$$\begin{aligned} [\partial_t + \Gamma - i\delta]\rho_{21} &= i\Omega(t)(\rho_{11} - \rho_{22})/2 \\ [\partial_t + \Gamma + i\delta]\rho_{12} &= -i\Omega^*(t)(\rho_{11} - \rho_{22})/2 \\ [\partial_t + \gamma]\rho_{22} &= i[\Omega(t)\rho_{12} - \rho_{21}\Omega^*(t)]/2 \\ \partial_t \rho_{11} &= \gamma\rho_{22} + i[\Omega^*(t)\rho_{21} - \rho_{12}\Omega(t)]/2 \\ \text{Tr}\{\hat{\rho}\} &= \rho_{11} + \rho_{22} = 1, \end{aligned} \quad (25)$$

where  $\Omega(t)=d\mathcal{E}(t)/\hbar$  is the time-dependent Rabi frequency ( $d$  is the dipole matrix element),  $\delta=(\omega-\omega_0)$  is the detuning of the central frequency  $\omega$ . The constants  $\gamma$  and  $\Gamma$  describe the relaxation rates of the excited state population ( $\rho_{22}$ ) and optical coherence ( $\rho_{12}, \rho_{21}$ ), respectively. Note that for pure spontaneous relaxation the condition  $\Gamma=\gamma/2$  takes place.

The most interest has a regime, in which the rep-

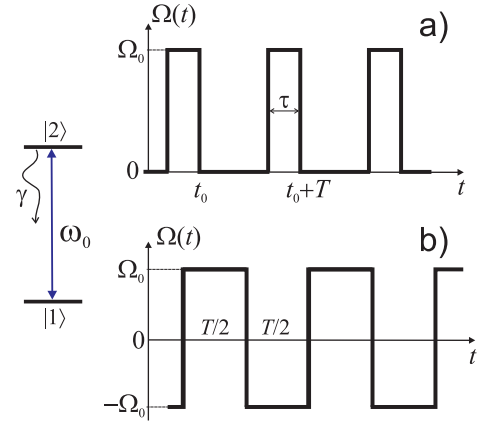


FIG. 5: Two-level system interacting with: a) periodic sequence of rectangular pulses; b) periodic sequence of  $\pm\pi$  phase jumps.

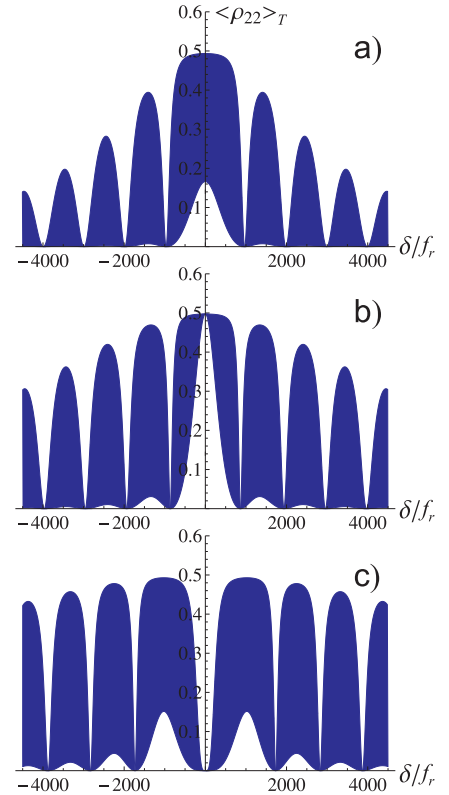


FIG. 6: The lineshape for direct frequency comb spectroscopy (see Fig. 5a) over large interval of detuning  $\delta$ , where the narrow resonances are visually inseparable (dark areas). Calculations are done for  $\Gamma=\gamma/2=0.02f_r$ ,  $\tau/T=0.001$  and for different pulse area: a)  $\Omega_0\tau=\pi/2$ ; b)  $\Omega_0\tau=\pi$ ; c)  $\Omega_0\tau=2\pi$ .

etition frequency significantly exceeds the relaxation rate of atomic system:  $\Gamma, \gamma \ll f_r$ , and also the realization of the condition  $\Omega_0\tau \geq 1$ . The Figs. 6a-c demonstrate the dependences of period-averaged value  $\langle \rho_{22} \rangle_T$  as a function of detuning  $\delta$  for different pulse area  $\Omega_0\tau=\pi/2, \pi, 2\pi$ . The structure of these lineshapes con-

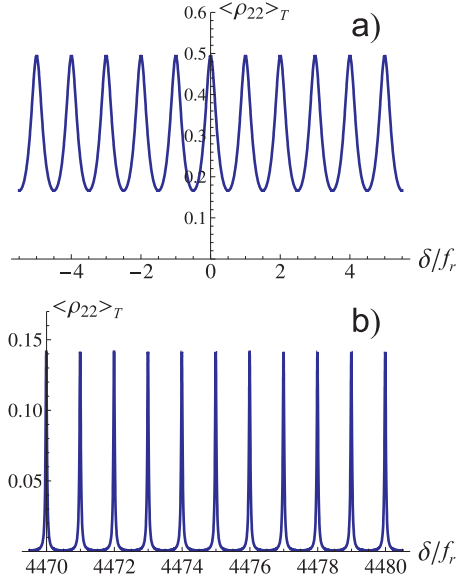


FIG. 7: The lineshapes for direct frequency comb spectroscopy (see Fig. 5a) over two small intervals of detuning  $\delta$ , where the individual Ramsey resonances are visually separated. Calculations are done for:  $\Gamma=\gamma/2=0.02f_r$ ,  $\tau/T=0.001$ ,  $\Omega_0\tau=\pi/2$ .

sist of large number of resonances at the frequencies  $\omega=\omega_0+m f_r$  ( $m=0,\pm 1,\pm 2,\dots$ ). Their amplitudes lie between two envelope curves. The dark areas in Fig. 6 correspond to the resonances, which are visually inseparable for given scale. More detail picture using small frequency intervals is presented in Fig. 7, where the individual resonances are visible. We have found that the lower envelope has a typical width of the order of  $2\Omega_0$  (see in Fig. 6) and it looks similar to the lineshape on  $\delta$  in the case of single-pulse Rabi spectroscopy. The upper envelope is much more broadened (with width of the order of  $\Omega_0 f_r/\gamma \gg \Omega_0$  in the case of  $\Omega_0\tau \geq 1$ ). Moreover, this envelope curve has a series of relatively narrow resonance-like dips (practically to zero), which can be characterized as quasi-forbidden spectroscopic zones. The positions of these zones are determined from the condition:  $\tau\sqrt{\Omega_0^2 + \delta^2} = 2k\pi$  ( $k=1,2,\dots$ ), and their widths are proportional to  $\gamma$ .

Note the high efficiency and simplicity of our method in the case of periodic pulse modulation. Indeed, for above considered task with rectangular pulses (see Fig. 5a) the operator  $\hat{A}(t_0 + T, t_0)$  includes the product of only two matrix exponents:

$$\hat{A}(t_0 + T, t_0) = e^{\tau \hat{L}(\Omega=\Omega_0)} e^{(T-\tau) \hat{L}(\Omega=0)}. \quad (26)$$

In result, the calculation of the dynamic steady-state in concordance with (15) becomes quite simple. Moreover, for two-level atom we have obtained the analytical expressions (exact and approximative) for the operator  $\hat{A}(t_0 + T, t_0)$  and vector  $\vec{\rho}(t_0)$ . In particular, for the short pulses ( $\tau \ll 1/\gamma$ ,  $\tau \ll T$ ) the following approximation takes

place:

$$\begin{aligned} \rho_{22}(t_0) \approx & \Omega_0^2 e^{\gamma T} (e^{2\Gamma T} - 1) \sin^2(\Omega\tau/2) \times \\ & \{ (e^{(2\Gamma+\gamma)T} - 1)\Omega^2 + 2e^{\Gamma T} (e^{\gamma T} - 1)\Omega\delta \sin(\delta T) \sin(\Omega\tau) - \\ & e^{\Gamma T} (e^{\gamma T} - 1) \cos(\delta T) [\Omega_0^2 + (2\delta^2 + \Omega_0^2) \cos(\Omega\tau)] + \\ & (e^{\gamma T} - e^{2\Gamma T}) [\Omega_0^2 \cos(\Omega\tau) + \delta^2] \}^{-1}, \end{aligned} \quad (27)$$

where  $T=(T-\tau)$  is a free evolution time between pulses,  $\Omega=\sqrt{\Omega_0^2 + \delta^2}$  is the generalized Rabi frequency. For the pure spontaneous relaxation ( $\Gamma=\gamma/2$ ) the expression (27) can be transformed to the form

$$\begin{aligned} \rho_{22}(t_0) \approx & \Omega_0^2 \exp(\gamma T/2) \sin^2(\Omega\tau/2) \times \\ & \{ 2\Omega^2 \cosh(\gamma T/2) + 2\Omega\delta \sin(\delta T) \sin(\Omega\tau) - \\ & \cos(\delta T) [\Omega_0^2 + (2\delta^2 + \Omega_0^2) \cos(\Omega\tau)] \}^{-1}. \end{aligned} \quad (28)$$

In addition, under condition of  $\tau \ll T$  we have a close approximation:

$$\langle \rho_{22} \rangle_T \approx \rho_{22}(t_0) [1 - e^{-\gamma T}] / \gamma T \quad (29)$$

with an absolute accuracy of small value  $(\tau/T) \ll 1$ . Thus, to reproduce with a good accuracy the lineshape of  $\langle \rho_{22} \rangle_T$  we can calculate only the dependence  $\rho_{22}(t_0)$  on  $\delta$ . Note that the Eq. (29) can be used for an arbitrary pulse form (not only rectangular).

In contrast, for frequency comb spectroscopy to numerically solve the Eq. (2) using the Fourier analysis we should use the following decomposition:  $\vec{\rho}(t) = \sum_m \vec{\rho}_m e^{m f_r t}$  ( $m = 0, \pm 1, \pm 2, \dots$ ), where the components  $\vec{\rho}_m$  satisfy the certain recurrent relations. As an example, the calculations of the dependencies in Fig. 6 require the using of more than  $10^5$  frequency components (for field and density matrix). It leads to the huge computational burden (comparable with supercomputer).

The last numerical example is shown in Fig. 8. There are the spectroscopy lineshapes for the field modulation, which is formed by  $\pm\pi$  phase jumps (see Fig. 5b). In the case of  $\Omega_0 \gg \gamma$ , these interesting dependencies have the lower envelope, which has a width of the order of  $2\Omega_0$  (see Fig. 8b,c,d).

Note that for free atom gas it is necessary to take into account the motion of atoms, which results in dependence of the density matrix on velocity  $\mathbf{v}$ . In this case the periodic steady-state  $\hat{\rho}(t+T, \mathbf{v}) = \hat{\rho}(t, \mathbf{v})$  should be found for each velocity group, and then the spectroscopic signal is computed with the use of velocity averaging. In addition, the performed in this article consideration of temporal periodic modulation can be adapted for the case of the spatially periodic modulation of the field parameters (amplitude, phase, polarization, etc.). It implies the proof of existence and the calculation of spatially periodic steady-state of the atoms in the spatial lattices formed by electromagnetic field (including the problems of laser cooling and trapping of atoms in optical lattices).

In conclusion, in the frame of density matrix formalism we have rigorously proven the existence theorem of



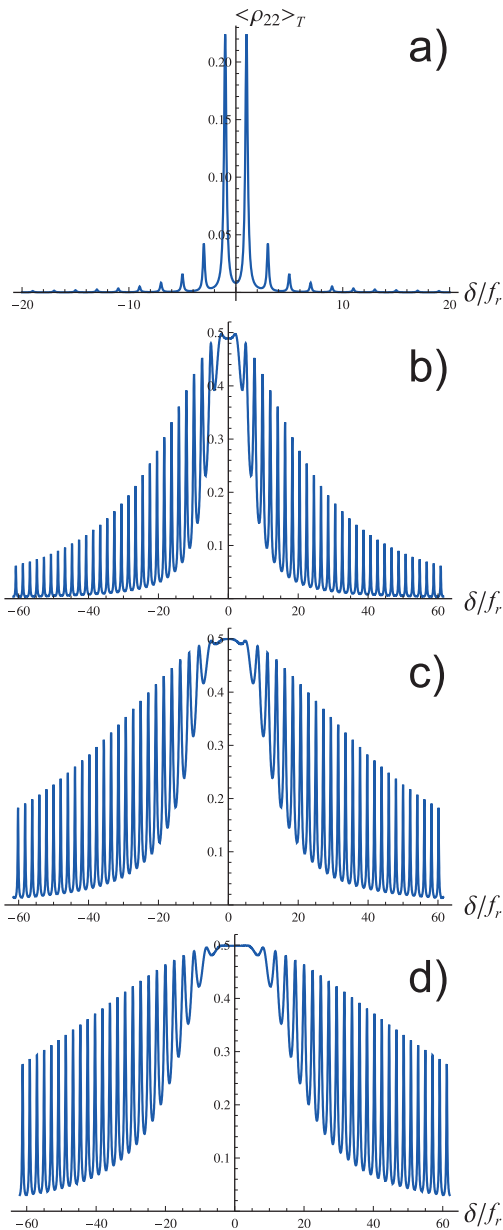


FIG. 8: The spectroscopy lineshapes for the field modulation, which is formed by  $\pm\pi$  phase jumps (see Fig. 5b). Calculations are done for  $\Gamma=\gamma/2=0.1f_r$  and for different Rabi frequency: a)  $\Omega_0=0.2f_r$ ; b)  $\Omega_0=5f_r$ ; c)  $\Omega_0=10f_r$ ; d)  $\Omega_0=15f_r$ .

the periodic steady-state for an arbitrary periodically-driven system. Due to the relaxation processes this state is realized as an asymptotics ( $t \rightarrow +\infty$ ) independently of initial conditions. The presented proof simultaneously contains a computational algorithm, which uses neither Floquet nor Fourier theories, i.e. our method automatically takes into account all frequency components. It radically simplifies the calculations for arbitrary types of periodic modulation (including the ultrashort pulses) and opens up great possibilities for analysis and development of new methods in the laser physics, nonlinear optics and spectroscopy. The importance of obtained results becomes especially obvious due to the infinite variety of possible periodic actions for different quantum systems.

We thank E. Demler for useful discussions. The work was supported by the Ministry of Education and Science of the Russian Federation (state assignment No. 2014/139, project No. 825), by the Russian Foundation for Basic Research (grants No. 14-02-00712, No. 14-02-00939, No. 15-02-08377, No. 15-32-20330). M. Yu. Basalaev was also supported by the Dynasty Foundation.

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